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Prediction Parameters of Radionuclide
Retention at Low-Level Radioactive Waste Sites

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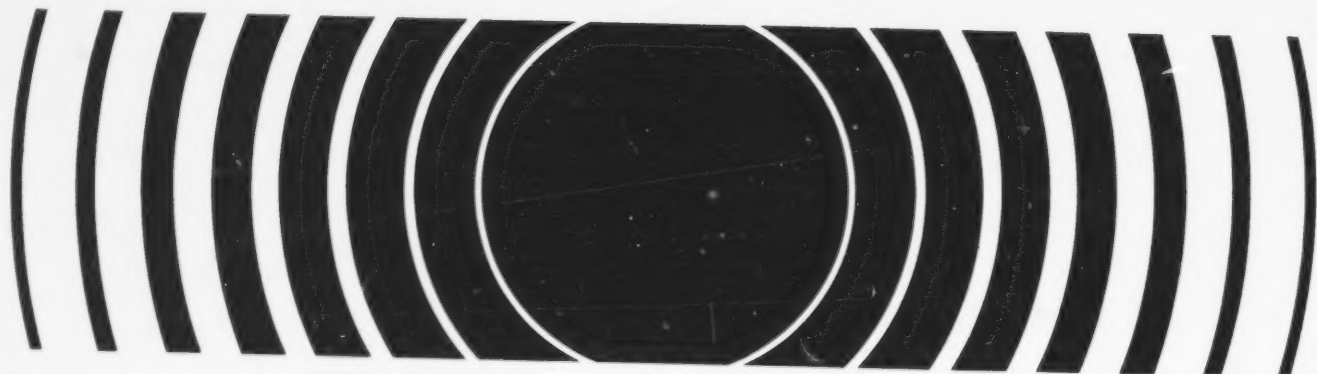
Office of
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Radiation

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16. ABSTRACT Three low-level radioactive waste (LLW) sites situated within three of the major surficial deposits of the United States are being investigated to determine predictive parameters that may exist for assessing the radionuclide retention at future LLW sites. Each of the major surficial deposits has a unique assemblage of sorptive minerals, texture, and chemical environmental factors that affect the sorption mechanism of the radionuclides. The LLW sites are located at Beatty, Nevada; West Valley, New York; and Barnwell, South Carolina. Preliminary results reveal predictive linear relationships for cesium and strontium in a plot of the percent sorptive minerals versus the sorption coefficient (Kd). The percent sorptive minerals are determined by a quantitative mineral method. Radionuclide sorption control mechanisms of cobalt, nickel, plutonium, uranium, americium, technetium, and iron reflect some retention related to mineralogy, but principal retention factors are more related to the nature and amount of organics, ligands, oxyhydroxides, redox potential, pH, and groundwater chemistry.		
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James Nelheisel

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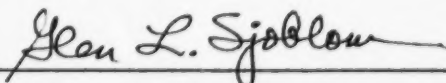
FOREWORD

The Low-Level Radioactive Waste Policy Act of 1980 makes each State responsible for disposing of low-level radioactive waste (LLW) either within the State or outside the State under compacts with other States. The States have requested, through the Presidentially-established State Planning Council on Radioactive Waste Management, that EPA provide a generally applicable environmental standard for the disposal of LLW. As a result of this request, EPA proposes to develop an applicable LLW standard in accordance with the authority and provisions of the Atomic Energy Act of 1954, as amended.

The EPA Office of Radiation Programs (ORP) plans to complete specific technical tasks during the development of the LLW standard. These tasks include waste classification and health risk assessments using model systems on a number of disposal options.

As part of the ongoing technical tasks, the Los Alamos National Laboratory, under Interagency Agreements with the Department of Energy, has assisted in providing laboratory and field data to establish a predictive methodology for States and State compacts to assess radionuclide retention at potential LLW sites. The methodology is based on data from three LLW sites situated in contrasting climates and surficial deposits. This report presents preliminary results obtained with the prediction methodology, which is a rapid, inexpensive method aligned with the capabilities of conventional laboratories. This predictive method, if fully developed, could replace the acceptable but more expensive sorption coefficient (Kd) method in the selection of suitable potential LLW sites.

The Agency invites all readers of this report to send any comments or suggestions to Mr. David E. Janes, Director, Analysis and Support Division, Office of Radiation Programs (ANR-461), U.S. Environmental Protection Agency, Washington, D.C. 20460.



Glen L. Sjoblom, Director
Office of Radiation Programs

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ABSTRACT

✓
The Environmental Protection Agency is evaluating site-specific soils from the Beatty, Nevada; Barnwell, South Carolina; and West Valley, New York, low-level radioactive waste (LLW) sites to determine critical parameters for predicting radionuclide retention. Each of the sites is located in one of the three major surficial deposits of the contiguous United States. These deposits have a unique assemblage of sorptive minerals, texture, and environmental factors that affect the sorption mechanism of the radionuclides. The sorption coefficients (K_d) of most radionuclides are highest in the alkaline Alluvial Basin deposits which contain montmorillonite and zeolites as principal sorptive minerals.

A plot of the percent sorptive minerals, determined by an applied quantitative technique, versus the sorption coefficient (K_d), reveals that a linear relationship exists for strontium and cesium in some of the deposits. Thus, strontium and cesium are predictable in relation to mineral type and quantity in a given chemical environment. Other radionuclides, however, reflect retention governed more by chemical factors such as pH, Eh (redox potential), presence of organics, ligands, and nature of the groundwater. Uranium, for example, shows high retention at the Barnwell site but low retention at the other two sites with the carbonate ligand in the groundwater probably being the main control factor. ↩

INTRODUCTION

The Low-Level Radioactive Waste Policy Act of 1980 requires that by 1986 the States dispose of their low-level radioactive waste within their State or outside the State under compact with other States. The Environmental Protection Agency (EPA) is developing numerical, generally applicable environmental standards under the authority of the Atomic Energy Act of 1954, as amended. During the standards development, EPA plans to complete specific technical tasks to use in assessing health risks of selected disposal options. As part of the technical tasks, in concert with health risk assessments, natural and engineered barriers are being evaluated at three existing low-level waste sites that have geologic media typical of the three major surficial deposits of the contiguous United States. This paper addresses the radionuclide retention capabilities of the natural barrier and presents a methodology for assessing potential radionuclide retention at future low-level radioactive waste (LLW) sites.

Soil has the ability to slow down the movement of radionuclides. Because of the many variables involved, this aspect has not been considered as an important factor in selecting waste sites. At present, laboratory measurements of site specific soils for sorption coefficients (K_d 's) of radionuclides is the most acceptable method of characterizing radionuclide retention, but this method is also very expensive and time consuming. In this method of analysis, the K_d of each radionuclide of concern is measured for its specific activity in the solid and liquid phase in controlled batch or column tests. The number generated as the ratio of the radionuclide retained on the solid divided by that in the liquid is the K_d of the radionuclide. The higher the K_d number, the greater the retention of the radionuclide. The K_d numbers vary in relation to several variables of which pH and type and amount of sorptive minerals are important; these factors can be measured with confidence by prescribed methods. Other factors are also important in control of radionuclide retention, and many of these are similar within similar surficial deposits.

The basic methodology prescribed in this report is to examine the Barnwell, South Carolina; Beatty, Nevada; and West Valley, New York, sites, each as a unique site within differing major surficial deposits. The textural, mineralogical, and chemical factors of each of the LLW sites within each of these surficial deposits is compared with the K_d measurements of radionuclides on site specific soils and groundwater from each of these sites to identify predictive parameters. The amount of sorptive minerals at each site is varied in quantity so that proportional increases of K_d may be related to type and amount of sorptive minerals.

A quantitative assessment of the mineralogy is required to determine the precise amounts of sorptive minerals in the respective samples for comparison with the Kd measurements. Preliminary results show that prediction of some of the radionuclides may be possible by careful mineral analysis.

The surficial deposits found at the three LLW sites cover large areas of the contiguous United States and are not confined to conventional physiographic boundaries. Since each deposit has a unique assemblage of minerals and soil and groundwater chemistry, a comparison of their capabilities to retard the migration of radionuclides also lends itself to considering the factors which control the sorption mechanisms of radionuclides.

The prediction method presented is tentative and requires validation. However, the methodology is less expensive and less time-consuming than the conventional Kd method for some of the important radionuclides and could be of value in selecting future LLW sites.

SURFICIAL DEPOSITS OF THE CONTIGUOUS UNITED STATES

The low-level radioactive waste (LLW) sites at Barnwell, South Carolina; Beatty, Nevada; and West Valley, New York, are located in unconsolidated surficial deposits which cut across physiographic provinces and occur in varying thicknesses above bedrock. Hydrologists have classified surficial deposits into surficial regions with similar physical properties, spatial locations, climate, and other factors relating to groundwater. The U.S. Water Resource Council (1968) has described the major areas of potential groundwater development and has delineated most of the major surficial deposits. Hunt (1974 and 1977) has described and mapped the geology of these surficial deposits. The surficial deposits shown in Figure 1 are those described by the the U.S. Water Resource Council (1968) with modification to include the New England glacial deposits by Hunt (1974).

Low Level Waste Sites

The LLW sites considered in this investigation are located in three different major surficial deposits which are present to some extent in 46 of the contiguous United States. West Valley, New York, is located in the Glaciated Central Region in transported glacially derived deposits of Pleistocene age; these constitute the youngest deposits. Barnwell, South Carolina, is situated in the Atlantic Coastal Plain in the unconsolidated marine sediments of Tertiary age. Beatty, Nevada, is located in the unconsolidated Alluvial Basin deposits of Tertiary age in the semi-arid to arid western States; these continental deposits are derived from the weathering of adjacent mountain ranges.

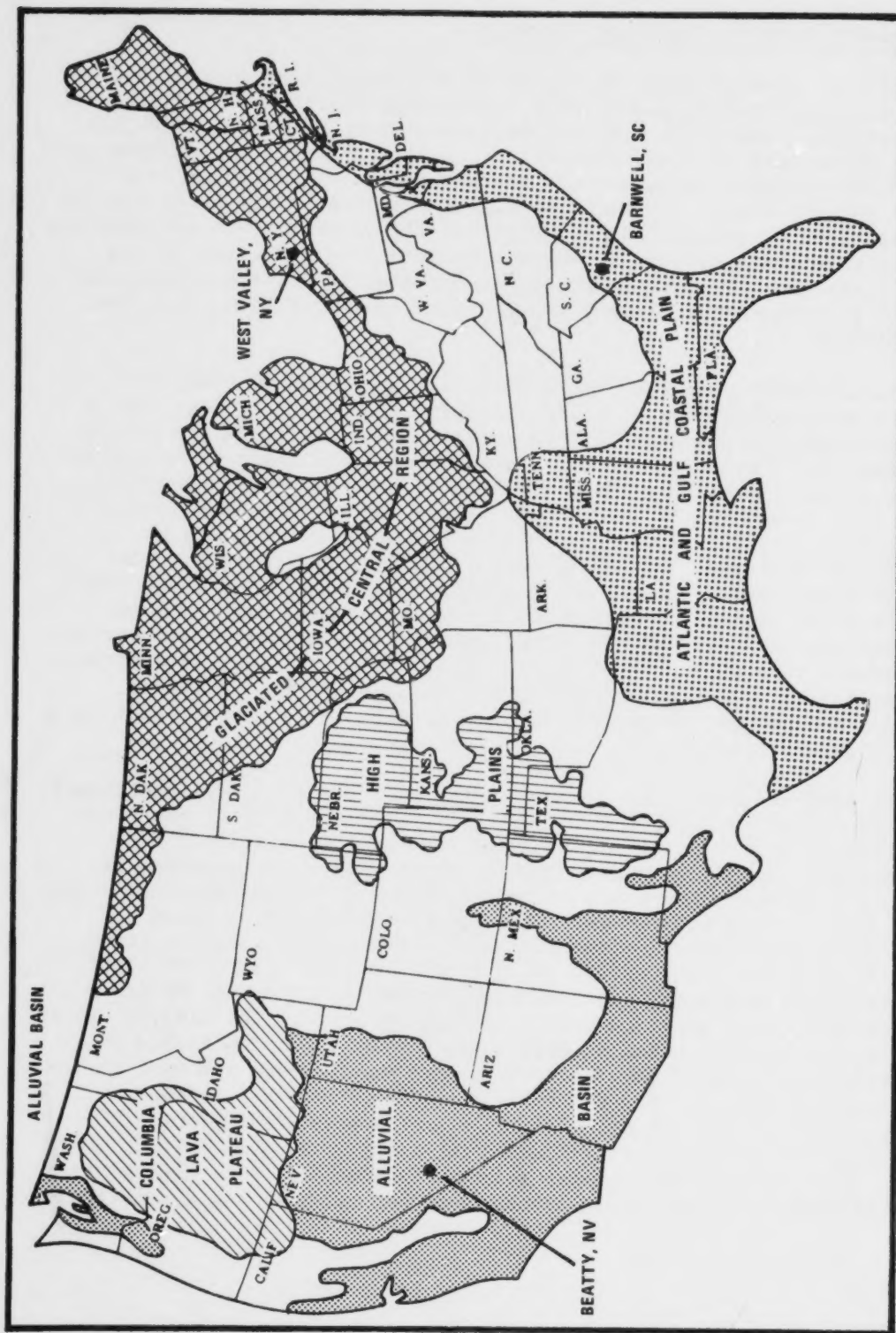


Figure 1. Surficial deposits of the contiguous United States and location of three commercial low-level radioactive waste sites. Source: U.S. Water Resource Council (1968) with modification to include glacial deposits of New England after Hunt (1974).

Clay Mineral Suite of Surficial Deposits

The clay mineral suite of the surficial deposits at each of the sites is related to its source rock, weathering conditions, and history of deposition. Since the clay minerals constitute the major sorptive mineral components of these deposits, their distribution in relation to climate and geologic factors could be expected to be reflected in a spatial pattern within the major surficial deposits. This study and the investigations of others indicate that the clay minerals and zeolites are the most sorptive materials as regards radionuclide retention by the common minerals. In general, the sorption potential of the common clay minerals from highest to lowest are montmorillonite, illite, chlorite, and kaolinite.

Millot (1979) discussed the general climatic control over clay minerals on a global basis. Montmorillonite (smectite) is most common from weathering of volcanic rock in poorly drained areas of the middle latitudes, while kaolinite occurs most abundantly in humid tropic zones where chemical leaching is heavy (Millot, 1979). Illite and chlorite appear most abundantly in the colder, higher latitudes. This general relationship is displayed in Figure 2 in the world ocean sediment distribution patterns (Neiheisel, 1983). Kaolinite is unique to the Atlantic Ocean because of the predominance of equatorial drainage from the intraplate regions of continents into a relatively smaller ocean basin and the lack of trenches which enables its transport to deposition sites across the basin. The sediment of the Pacific Ocean, by contrast, is dominated by a montmorillonite clay-mineral suite in the middle latitudes and equatorial regions and a chlorite-illite clay-mineral suite in the high latitudes.

The clay-mineral suites of the surficial deposits of the continents have a distribution pattern that generally parallels that of the ocean basins (Figure 2). The illite-chlorite clay-mineral suite is characteristic of The Glaciated Central Region which is comprised of materials transported by the continental glaciers of the Pleistocene age from colder, northern latitudes. The montmorillonite-rich loess deposited by the prevailing westerly winds constitutes sizeable local surficial features over considerable portions of the glacial tills of the Glaciated Central Region; however, the predominant clay-mineral suite of this region is best characterized as illite-chlorite. The Alluvial Basin deposits of the western United States are comprised predominantly of montmorillonite derived from weathering of volcanic rock abundant in the Cretaceous and Tertiary age deposits of that region. Kaolinite is characteristic of the Atlantic Coastal Plain, and montmorillonite constitutes the principal clay mineral of the Gulf Coastal Plain (Figure 2). The kaolinite of the Atlantic Coastal Plain is derived from the weathering of the adjacent Appalachian System which has a relatively

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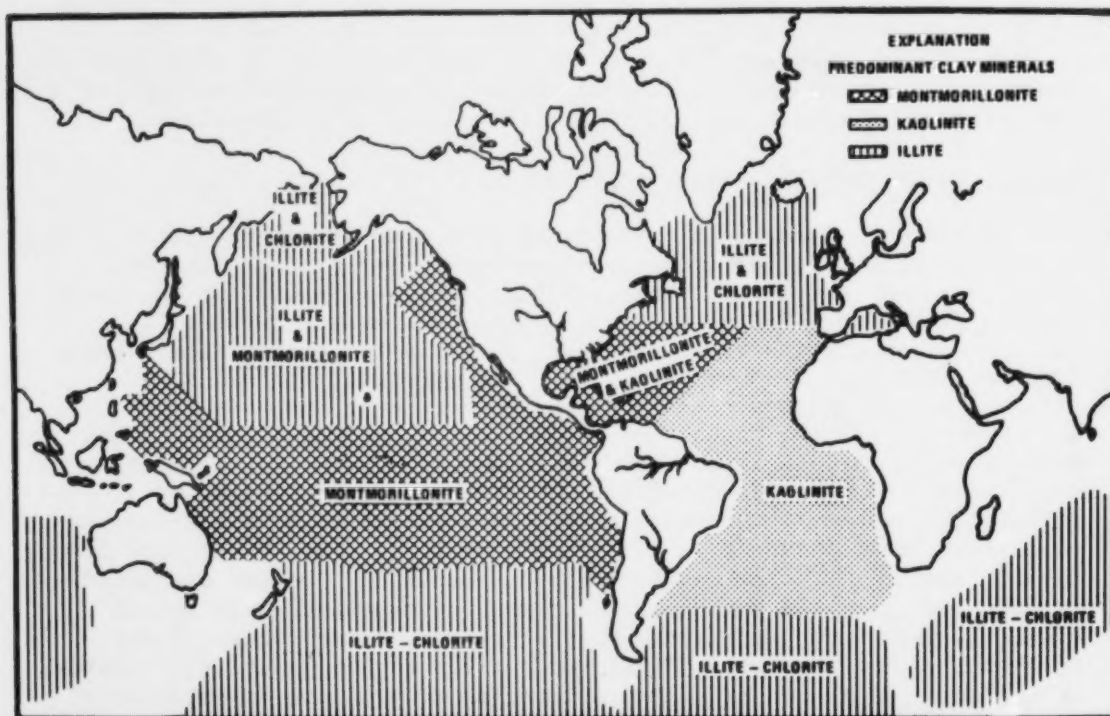
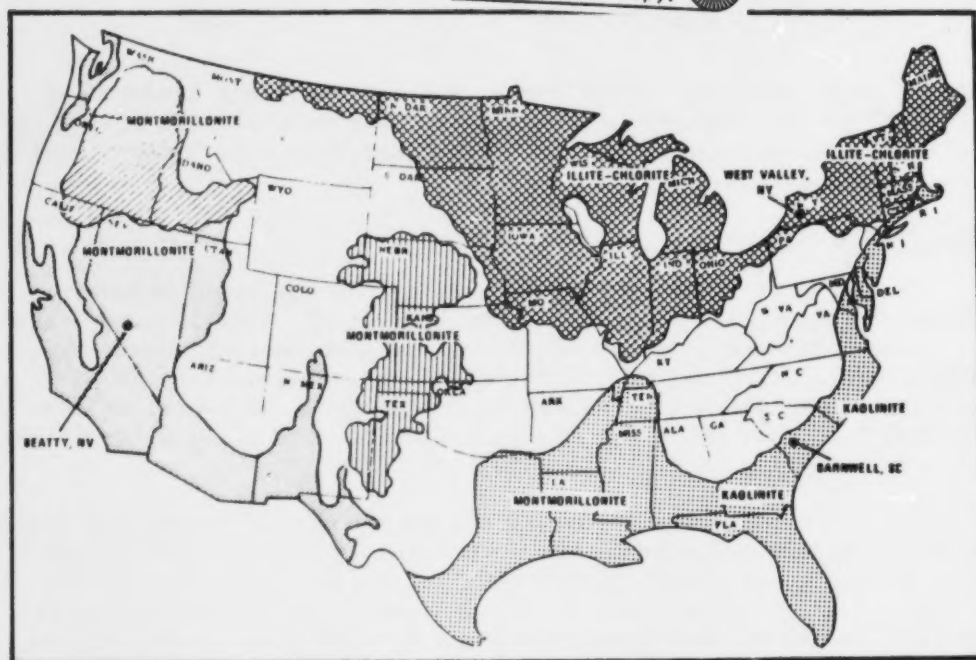


Figure 2. Comparison of clay-mineral suite of major surficial deposits of the contiguous United States and world ocean basins.

warm climate, granitic source rocks, and acid leaching environment favorable to the formation of kaolinite. The Gulf Coastal Plain, by contrast, contains clay minerals transported from the montmorillonite-rich western States.

Chemical Aspects of the Surficial Deposits

The chemical environmental factors of the surficial deposits influence radionuclide retention. Onishi, et al. (1981), have listed pH, Eh, cation exchange capacity (CEC), competitive ions, presence of oxyhydroxides, and presence of organics and inorganic ligands as important chemical factors affecting the sorption mechanism of some of the radionuclides. Of those chemical factors, the pH is perhaps the least variable and most highly significant.

The pH of the Atlantic Coastal Plain and Glaciated Central Region is generally acidic and encompasses a pH range of 4 to 7. The Alluvial Basin deposits, by contrast, are alkaline with the pH range between 7 and 9. The alkaline pH is a result of evaporation of calcium carbonate deposits formed in the unsaturated zone of semi-arid to arid regions. Swanson (1982), in an investigation of the stability of nickel and cobalt in Hanford soil, demonstrated that the retention capability of these radionuclides is higher in the alkaline pH (9.0) as compared to an acid pH (6.0). Thus, one might expect greater retention of radionuclides in the Alluvial Basin deposits based on the pH alone for some of the radionuclides. However, the most highly sorptive minerals also occur in these deposits, and radionuclide retention is predictable for some of the radionuclides that are specifically controlled by the mineralogy.

Background Terrestrial Radiation Levels

Data on the background terrestrial radiation levels of United States surficial regions in the vicinity of nuclear power plants have been compiled by Oakley (1972). This information, extrapolated to the total surface area of the United States, revealed the lowest terrestrial background reading in the Atlantic and Gulf Coastal Plain regions (mean dose equivalent of 22.8 mrem/y). The northern central areas had intermediate background radiation levels (mean dose equivalent of 45.6 mrem/y), and the highest readings were in the Denver area of the Rocky Mountain region (mean dose equivalent of 90 mrem/y). The Alluvial Basin surficial deposits apparently have a background reading of intermediate range if the Reno, Nevada, area is cited as typical (dose equivalent of 45.6 mrem/y). More recent airborne radiometric data from the DOE National Uranium Resource Evaluation (NURE) program conducted between 1976 and 1982 could provide total coverage and could update present values. The background terrestrial radiation values of the surficial deposits to some degree reflect the mineralogical and chemical environments of these units and may provide correlative information of value in defining radionuclide retention.

FACTORS GOVERNING RADIONUCLIDE RETENTION

Texture

Korte, et al. (1976), have determined that the clay content (less than 2 micron size) of a soil was the more useful parameter of several considered in determining retention or attenuation of a particular element in several soil types. The parameters considered included cation exchange capacity, pH, surface area, free iron oxides, total Mn, soil texture, soil class, and cursory mineral analysis. Gee and Campbell (1980), in an analysis of two Hanford, Washington, soils in which the only difference between the soils was the amount of clay-size materials, found radionuclide retention greatest in the sample with the higher clay content. Several other investigations leave little doubt that the most sorptive minerals are in the clay-size fraction. Thus, a first approximation of radionuclide retention might be made on a standard texture grain-size accumulation curve that gives weighted amounts of sand, silt, and clay-size fractions.

Ames and Rai (1979) published summary information of strontium and cesium retention on pure mineral phases. Quartz and feldspar, more common to sand and silt-size fractions, have low retention (K_d) values compared to the common clay minerals of clay-size fractions (Table 1).

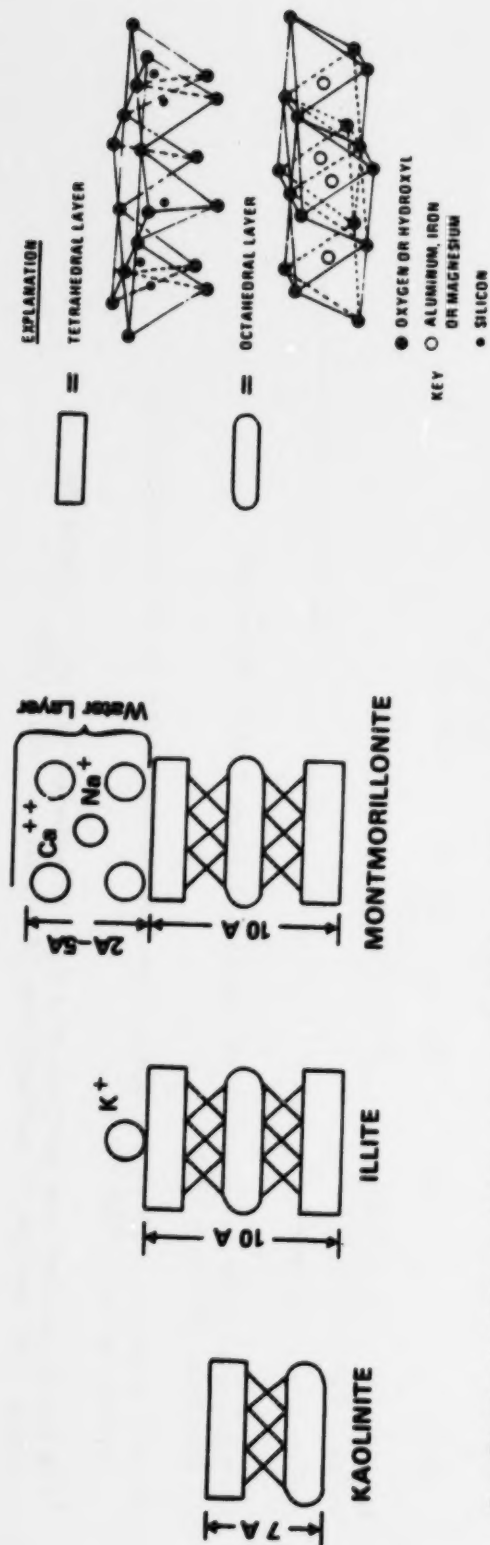
Montmorillonite, the common clay mineral with the greatest radionuclide retention, occurs in the smallest size range of the common clay minerals; average kaolinite, illite, and montmorillonite sizes are, respectively, 1.0, 0.3, and 0.1 microns in the direction of the larger average dimension (Figure 3). Montmorillonite, with smallest particle size, has the largest surface area per unit volume. The correlation of surface area to clay mineral species is apparent from measurements of soils at several locations in two separate investigations (Table 2). If the surface area is divided by the percent clay fraction for the predominant clay mineral species, it is apparent that the ratio for kaolinite approximates 1.5, whereas the ratio for the montmorillonite-rich clay size approximates 5 on several samples reported by Korte, et al. (1976); this is also apparent in the data presented by Pietrzak, et al. (1981). Illite has a ratio between that of kaolinite and montmorillonite which correlates with the particle size data depicted in Figure 2. Thus, we see that texture and surface area reflect the mineral composition.

Sorptive Minerals

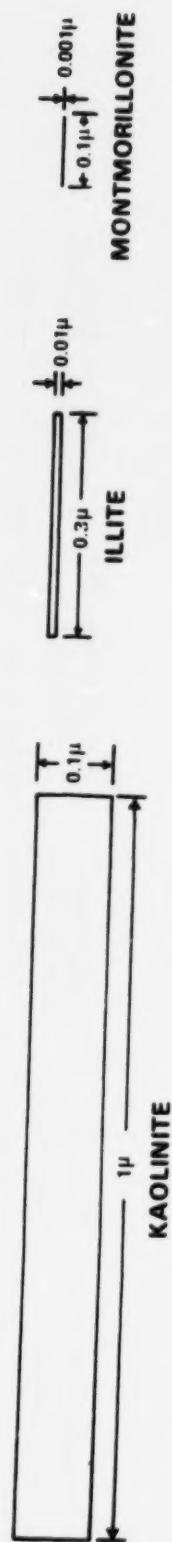
The term sorption, as used in this investigation, includes adsorption and absorption of radionuclides on solids from an aqueous solution. For cesium and strontium, the clay minerals having the highest sorptive characteristics, in increasing order, are kaolinite, illite, and montmorillonite (Table 1). This relative order is also generally similar for other radionuclides for which cation exchange is an important sorption mechanism. The sorption potential of the clay minerals for a radionuclide is related to a number of factors, including their charge and structural and chemical characteristics.

Table 1. Typical Kd Values of Strontium and Cesium on
Pure Mineral Phases Characteristic
of Sand, Silt, and Clay-Size Materials

<u>Texture</u>	<u>Minerals</u>	<u>Distribution Coefficient (Kd)</u>	
		<u>Strontium</u>	<u>Cesium</u>
Sand-Size	Quartz	0 - 5	0
	Feldspar	9	9
	Mica	5	5 - 15
Silt-Size	Quartz	0 - 5	0
	Feldspar	9	9
	Mica	5	5 - 15
Clay-Size	Kaolinite	15	45
	Illite	100	400
	Montmorillonite	150	4900



A - Structural thickness of three common clay minerals in angstrom units (A)



B - Comparative particle size of common clay minerals in micron units in natural deposits

Figure 3. Chemical structure and comparative particle size of common clay minerals in the surficial deposits.

Table 2. General Relationship Between Texture, Surface Area,
and Clay Mineral Composition

Number of Soil Samples	Texture (% Clay-Size)	Predominant Clay Mineral	Surface Area (a) (m ² /g)	Surface Area % Clay-Size Fraction
2(b)	4-5	Kaolinite	8-9	2
3(b)	32-62	Kaolinite	51-67	1-2
1(b)	11	Illite	38	3
4(b)	35-49	Montmorillonite	121-126	5
Barnwell(c) Coastal Plain	15	Kaolinite	13	1

(a) Surface area by ethylene glycol monoethyl ether (EGME) technique (Heilman, et al., 1965).

(b) Data from 10 of 11 soil samples from various locations in the United States (Korte, et al., 1976).

(c) Barnwell texture and surface area from Brookhaven National Laboratory using EGME measure of surface area (Pietrzak, et al., 1981).

The clay mineral structures of kaolinite, illite, and montmorillonite are depicted in Figure 3. Kaolinite is made up of one tetrahedral sheet and one octahedral sheet with the sheets strongly held together by hydrogen binding. Kaolinite particles have a small negative surface charge, and sorption is believed to be mainly along broken bonds at the edges of the crystal structure.

Illite consists of one octahedral sheet between two tetrahedral sheets (Figure 3). The charge density of illite is the highest of the three clays considered, but due to its neutralization with non-exchangeable potassium ions, the cation exchange capacity is relatively low. Sorption is largely on external surfaces of this clay mineral.

Montmorillonite clay is the more common variety of the smectite clay minerals. It has a layer structure similar to illite, but, in addition, has one-to-two layers of water between tetrahedral sheets, with thickness depending on the variety of cationic species in the water interlayer (Figure 3). Sorption occurs on both the surface and in the water interlayer system. Montmorillonite has an intermediate charge but has the highest sorption and cation exchange capacity of the clay minerals because of the unusual intercalation properties and small particle size (less than 2 microns).

The zeolite clinoptilolite occurs in fair abundance in the Alluvial Basin deposits. Sorption measurements by Daniels, et al. (1982), on the clinoptilolite of Yucca Mountain tuffs for cesium indicate a K_d of 3.8×10^4 ml/g; strontium has generally similar sorption ratios for this zeolite. This mineral ranks with montmorillonite in sorption potential for many of the radionuclides. The measurement of clinoptilolite and other zeolites, therefore, is important in evaluating radionuclide retention potential of minerals.

The presence of quartz, feldspar, and mica, common in the sand and silt-size fractions of the surficial deposits, is considered to contribute little to the sorption of the radionuclides. Since sorbent minerals of clay size occur as coatings on some of the sand- and silt-size minerals, radionuclide retention is significant in larger size fractions because of the sorptive clay coatings, as will be shown later. The values listed in Table 1, for example, are for pure mineral phases free of adhering sorptive minerals of clay size.

Chemical Factors

Some of the chemical factors that may govern radionuclide retention include pH, Eh (redox potential), cation exchange capacity, and the presence of ligands, organics, and oxyhydroxides. The cation exchange capacity is a useful correlation to clay mineral composition, but according to Korte, et al., 1976, this parameter does not necessarily improve the ability to predict the movement of cations through the

natural soils. The cation exchange capacity is expressed in milliequivalents per 100 grams. Average values for pure mineral phases are 10 for kaolinite, 15 for illite, and 100 for montmorillonite. While cation exchange capacity is not as significant for most radionuclides, it is, however, considered especially significant to strontium, cesium, plutonium, and americium (Onishi, et al., 1981).

The pH is probably the most important chemical factor controlling the sorption of some of the radionuclides. Within the surficial deposits, this value is generally within a fixed range and given more test data, predictions based on this parameter are a foreseeable possibility for some radionuclides. The alkaline nature of the western soils is related to caliche formation in the semi-arid to arid climate, while the soils of the humid eastern United States are generally acid in nature. The marked change in radionuclide retention with change in pH demonstrated by Swanson (1982) for nickel and cobalt in a Hanford soil was discussed in an earlier section.

The Eh (redox potential) in the surficial deposits is oxic in nature. However, reducing conditions may occur in some trenches of LLW containing high concentrations of organics or oxygen depletion factors that result in a depleted quantity of oxygen such as reported by Weiss and Colombo (1980) for the West Valley site. As shown by Onishi, et al. (1981), the radionuclides most sensitive to such change are technetium, uranium, neptunium, and plutonium. Technetium, for example, has low retention under oxic conditions but moderate retention under reducing conditions.

The chemical factors within the surficial deposits are more difficult to predict because of a lack of understanding regarding their physicochemical nature or their predictable occurrence within the surficial deposits. Taking uranium as an example, Sherwood and Serne (1983) show that the retention of this nuclide is very sensitive to pH, oxidation state (Eh), the amount of complexing carbonate or sulfate, and organic ligands. The presence of high carbonate in the groundwater has the potential of rapid mobilization of uranium as anionic uranyl complexes; with relatively low carbonate in the groundwater, the uranium is less mobile and will be retained in larger quantities by the soil.

Since several sorption mechanisms may operate simultaneously for some radionuclides, the chemical control factors are difficult to assess for prediction of radionuclide retention. Their use for predicting radionuclide retention becomes increasingly greater by comparisons of chemical differences existing between major surficial deposits of the contiguous United States. Within the surficial deposits, comparisons of retention of radionuclides and chemical factors, with fewer variables involved, may eventually highlight the principal factors for use as predictive parameters. Laboratory tests may then define the range over which the particular chemical factor operates.

METHOD OF INVESTIGATION FOR PREDICTIVE PARAMETERS

A method to determine the potential use of textural, mineralogical, and chemical factors to predict radionuclide retention involves the following:

- (1) Laboratory measurements of sorption coefficients (K_d) of radionuclides of interest on site-specific samples and with site-specific groundwater. This constitutes the most acceptable state-of-the-art measure of radionuclide retention and will be used as a means of comparison with other measurements in identifying predictive parameters.
- (2) Fractionating samples while retaining chemical integrity to vary the amount of sorptive minerals in each sample. This is necessary to identify linear relationships that may exist if the percent sorptive minerals are compared with the sorption coefficients of radionuclides.
- (3) Quantitative evaluation of sorptive minerals to establish precise amounts in samples.
- (4) Measurement of background radiation and chemical properties of soil and groundwater believed significant to radionuclide retention.

Sorption Coefficient (K_d) Measurements

The philosophy, methodology, and applications of sorption measurements are discussed in detail in the Los Alamos National Laboratory investigation of the Yucca Mountain tuffs by Daniels, et al., (1982). In this investigation using similar methods, site-specific soil samples from the Beatty and Barnwell sites from 6- and 12-meter depths were prepared with site-specific groundwater spiked with radionuclides introduced at the Los Alamos Laboratory. Groundwater with the experiments were prepared with (1) ^{59}Fe , ^{60}Co , ^{85}Sr , ^{131}I , and ^{137}Cs , (2) ^{63}Ni , (3) ^{95}mTc , (4) natural uranium, (5) ^{239}Pu , and (6) ^{241}Am using the appropriate groundwater (Wolfsberg, et al., 1983). The pH values (± 0.5 unit) were ~ 8.7 and ~ 6.0 for the Beatty and Barnwell waters, respectively. The group of mixed nuclides and ^{95}mTc were analyzed by gamma counting on a GeLi detector. A NaI detector was used for ^{241}Am . Uranium was determined by neutron activation and delayed-neutron counting, and ^{239}Pu and ^{63}Ni were determined by liquid-scintillation counting. All sorption measurements were for 6 weeks contact time, and no desorption measurements were performed in this study (Wolfsberg, et al., 1983).

Some measurements of sorption were performed on samples in a controlled atmosphere of nitrogen containing <0.2 ppm oxygen and <20 ppm carbon dioxide. The purpose of these measurements was to investigate possible changes in sorption caused by any reducing minerals in the soils. Lower carbonate concentrations (due to loss of CO₂) in the controlled atmosphere may also affect sorption of some elements. The pH values of the waters in the controlled atmosphere were 0.5 to 1.0 units higher than in air (Wolfsberg, et al., 1983).

The sorption ratio is defined by

$$K_d = \frac{\text{activity on solid phase per unit mass of solid}}{\text{activity in solution per unit volume of solution}}$$

The K_d is the measure of the specific activity in the soil phase to that of the specific activity of the liquid phase for the radionuclide of interest in the controlled batch tests. The higher the K_d number, the slower a radionuclide will migrate through the soil relative to groundwater.

Fractionation of Samples

Knowing that minerals with the greatest sorption potential naturally occur in the finer grain size, the samples were fractionated to study whether linear relationships exist between proportional amounts of sorptive minerals and radionuclide retention. The soils reported in this preliminary investigation were disaggregated and separated texturally by sieving techniques (Wolfsberg, et al., 1983). The disaggregating techniques used did not alter the chemical properties. A more vigorous treatment, such as used in determining particle size distributions, would have given better textural separations; however, dispersants were avoided and extremes in agitation were not used to maintain chemical integrity of the samples. The fractionated samples are (1) a predominant sand-size fraction; (2) a silt-size fraction, and (3) a mixed silt- and clay-size fraction. Some of the clays in these fractions are apparently strongly mechanically bonded to larger mineral particles. Clays in the Barnwell samples are largely aggregates of kaolinite clay; Keller (1982) discusses the unusual nature of this clay mineral which occurs as aggregates in silt-size as well as typically clay-size materials.

The division of fraction sizes used in this investigation is in accordance with U.S. Department of Agriculture specifications. In this grouping, gravel represents particles greater than 2 mm size; sand represents particles between 2 mm and 50 micron-size; silt represents particles between 50 micron-size and 2 micron-size; clay represents particles less than 2 micron-size. Gravel-size particles were removed prior to testing.

Quantitative Mineral Analysis

The highly sorptive minerals occur mainly in the clay-size fraction, and some are aggregated in the silt-size fraction. It is therefore important to use a mineralogical identification method that is as quantitative as possible to obtain accurate information on weight percent in the sample. Several quantitative techniques to identify clay minerals have been described by investigators and have been evaluated by Pierce and Siegel (1969). While all of the methods have merit, some are more thorough in addressing all the mineral phases in the sample. The technique we used is designed to assess, as quantitatively as possible, the percent of sorptive minerals in the total sample.

The method employs separate x-ray diffraction analysis of representative portions of both the silt- and clay-size fractions to eliminate some of the minerals that, if present in a combined silt- and clay-size fraction, would cause interference peaks on the resulting diffractogram. Thus, clay minerals are predominant in clay-size fractions (except for kaolinite at the Barnwell site) and quartz and feldspar dominate the silt-size fraction. By preparing a standard grain-size accumulation curve for a sample, the weight of each size-fraction is determined for use in calculation of the average of each mineral in the total sample (Figure 4).

The Los Alamos Laboratory performed mineral analysis on the first samples tested for the Barnwell and Beatty sites using representative and fractionated samples. Samples were ground to a powder in acetone and packed into glass holders. Clay samples on filter paper were suspended in deionized water and sedimented onto glass slides to form oriented aggregates (Wolfsberg, et al., 1983). X-ray diffraction was performed using a Siemens D500 Diffractometer providing Cu K-alpha radiation. Each sample was run from 2° to 32° two-theta. Diffraction data were collected and processed by the Siemens DIFFRACC V system. The system identified X-ray diffraction peaks and calculated their d-spacing and integrated intensities (Wolfsberg, et al., 1983).

Mineral identifications were made by comparison of sample X-ray diffraction patterns with patterns from the JCPDS mineral file. Identifications were also confirmed by petrographic examination, where possible. Estimates of mineral percentages for each of the size-fractions were made by a combination of methods that included use of X-ray diffraction intensity peak correction (Hubbard, et al., 1976) and comparison of sample patterns to patterns for materials of known mineral composition (Wolfsberg, et al., 1983):

The sand-size fraction was analyzed by standard petrographic techniques using at least a statistical 300-grain count. Samples were also tested by x-ray diffraction techniques for determining minor amounts of sorptive clay and alteration or weathering products associated with the sand-size particles.

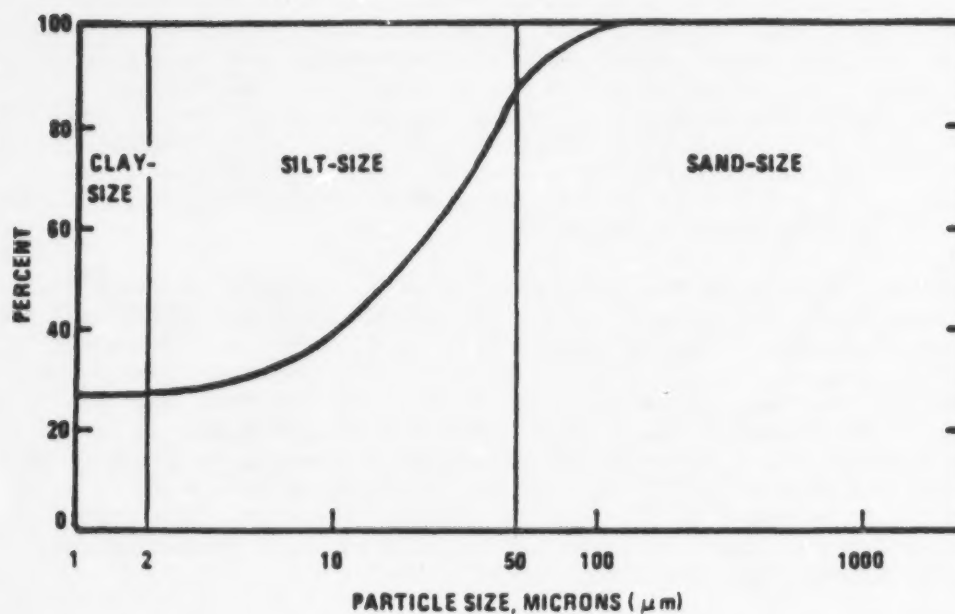
QUANTITATIVE MINERAL DETERMINATION

CALCULATION

PRODUCT OF FRACTIONAL COMPOSITION
AND GRAIN SIZE

PROCEDURE

● GRAIN-SIZE DETERMINATION



- MINERAL EVALUATION OF SIZE-FRACTIONS
 - SAND-SIZE – PETROGRAPHIC ANALYSIS
 - SILT-SIZE – X-RAY DIFFRACTION ANALYSIS
 - CLAY-SIZE – X-RAY DIFFRACTION ANALYSIS

Figure 4. Method for determining percent sorptive minerals in samples from LLW sites.

The percentage of each mineral in the sample is ideally calculated as the sum of the fractional percentages of the minerals in the sand, silt, and clay-size fractions.

Chemical Factors

The chemical factors tested for in this investigation by Los Alamos Laboratory included the pH, Eh, CEC, natural radioactivity, water analysis of site-specific groundwater, and soil analysis. The pH and Eh were determined in the field. Natural radioactivity of the soil was determined by a GeLi detector and by a liquid scintillation counter for water (Wolfsberg, et al., 1983). These values and nonradioactive isotopes or radionuclides of interest were determined and the values applied as specified by Gillham, et al. (1980), in computing the sorption coefficient.

COMPARISON OF RADIONUCLIDE RETENTION IN SURFICIAL DEPOSITS

The average sorption coefficients (Kd's) of radionuclides at the three LLW sites are shown in Figure 5. The Kd values for the Beatty and Barnwell sites are from laboratory determinations using site-specific soil and groundwater in tests conducted by the Los Alamos National Laboratory (Wolfsberg, et al., 1983). Data for the West Valley, New York, site is from an investigation by Weiss and Colombo (1980) of Brookhaven National Laboratory in which site-specific soils and trench water were used. Tests are still in progress at the Los Alamos Laboratory for the West Valley site.

As anticipated, negligible retention of tritium, carbon-14, iodine, and technetium occurs at any of the sites since these elements are present in the case of tritium as water or as anionic species or covalently bonded compounds. The greatest overall retention of radionuclides is in the Alluvial Basin deposits which contain an alkaline pH due to the calcium carbonate (caliche formation) and highly sorptive montmorillonite and zeolites. Even though the quantity of sorptive minerals is less at the Beatty site, the types of sorptive minerals (montmorillonite and zeolites) have higher sorption capabilities than those of the other surficial deposits. The alkaline pH of the Alluvial Basin, as compared to the acidic pH of the other deposits, is undoubtedly also a major contributing factor since solubility of many of the radionuclides is increased by the more acidic condition.

The highest retention for uranium occurs in the Coastal Plain sediments at the Barnwell site. Uranium retention is an order-of-magnitude higher at Barnwell than the other locations, probably due to the lower carbonate ion content in the groundwater at the Barnwell site (Wolfsberg, et al., 1983).

LLW SITE	SURFICIAL REGIONS & CLAY MINERAL SUITE	pH	RADIONUCLIDES – OXIC CONDITIONS										
			3H	129I	99Tc	238U	90Sr	63Ni	60Co	137Cs	239Pu	55Fe	241Am
WEST VALLEY, NY	GLACIATED CENTRAL ILLITE CHLORITE	6.8	1	1	1	1	1	1	2	3	3	3	
BARNWELL, SC	ATLANTIC COASTAL PLAIN KAOLINITE	5.9	1	1	1	2	2	2	4	4	4	5	
BEATTY, NV	ALLUVIAL BASIN MONTMORILLONITE ZEOLITES	7.3	1	1	1	1	2	3	4	3	3	5	



Figure 5. Average radionuclide retention at three low-level radioactive waste sites located in major surficial deposits in the United States.

The retention rates for cobalt and nickel are higher by more than an order-of-magnitude at the Beatty site in the Alluvial Basin deposits than at the other two sites. This is probably a function of alkaline pH rather than a complexing agent. Tewari, et al., 1972, explains the marked increase of cobalt on MnO_2 for the pH between 5.5 and 8.0 as a result of hydrolysis. Thus, this effect may explain the higher cobalt retention at the Beatty site compared to that at the West Valley or Barnwell sites (Figure 5).

Mineralogy can be highly significant in the assessment of the radionuclide retention of cationic species such as strontium and cesium. Both strontium and cesium show site specific correlation of sorptive mineralogy and K_d . The behavior of these nuclides may be predictable if one uses quantitative mineralogical techniques.

TEXTURE AND MINERAL COMPOSITION IN RELATION TO K_d VALUES

The triangular textural diagram depicted in Figure 6 shows the quantities of sand-, silt-, and clay-size fractions determined from the standard grain-size accumulation curve of the samples from the three LLW sites. The clay-size fraction ranges from 7 to 15 percent at the Beatty site, and 21 to 36 percent at the Barnwell site. The one sample tested at the West Valley site contained 33 percent clay-size material.

The texture, mineral composition, and sorption coefficient of radionuclides on samples from trench depth at the Beatty and Barnwell sites are listed in Tables 3 and 4. At each site it is generally apparent that the retention of many of the radionuclides increases with increased clay-size fraction; however, there are other complicating factors that govern simple proportional responses with most of the radionuclides. The texture of site-specific soils, however, does provide a rough index of relative retention for some of the radionuclides.

An assessment of the highly sorptive minerals in a sample is more meaningful than the percent clay-size fraction discussed previously if the quantitative method is used to obtain mineral percentages. Inspection of the texture versus highly sorptive minerals listed in Tables 3 and 4 shows general correlation of the clay-size fraction and the sum of the montmorillonite and zeolite (clinoptilolite) at the Beatty site. However, this relationship is not apparent for the Barnwell samples; the amount of sorptive kaolinite exceeds the clay-size fraction by a considerable amount. The kaolinite in the Barnwell samples occurs in abundance in all size fractions. While most clay minerals occur in greatest abundance in the clay-size fraction, kaolinite is more diverse in occurrence. Keller (1982) describes the wide range in textural patterns of kaolinite ranging from strictly clay-size to aggregates of silt-size and even larger sizes as a function of its mode of origin in varieties of geochemical environments. The kaolinite in the Barnwell samples calculated from the quantitative mineral evaluation method are 45, 64, and 80 percent in sample numbers 4, 12, and 14, respectively (Table 3).

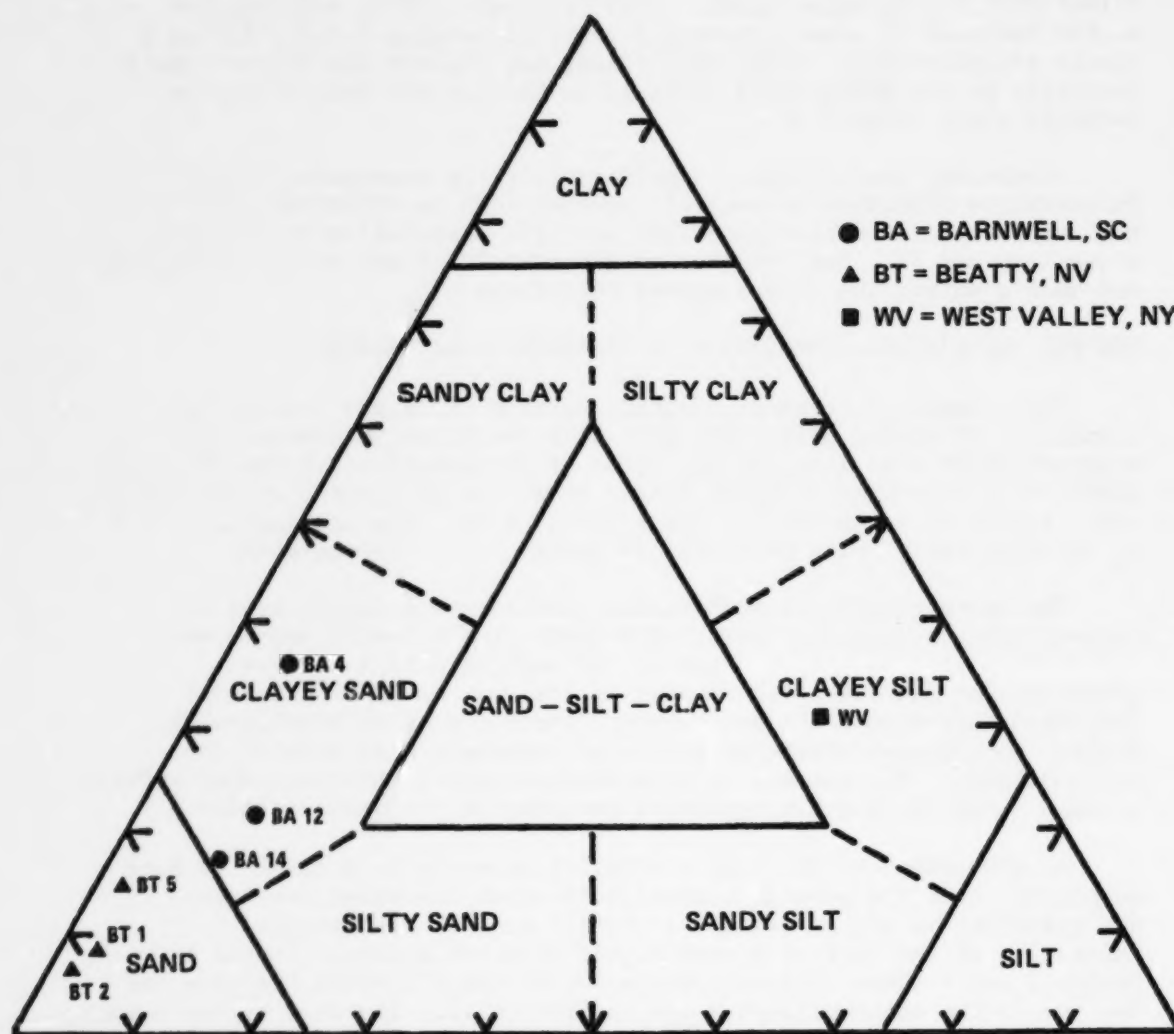


Figure 6. Ternary textural diagram of sand - silt - and clay-size materials at three low-level radioactive waste sites.

Table 3. Texture and Mineral Composition of Soil Samples from Beatty and Barnwell
Low-Level Radioactive Waste Sites

Sample Number	Texture			Mineral Composition				
	Sand	Silt	Clay	Quartz	Feldspar	Mica	H.M.	Kaolinite Montmorillonite Zeolite
Beatty								
1	89	3	8	15	80	-	-	3
2	91	2	7	13	80	1	-	3
5	83	2	15	8	80	2	-	7
Barnwell								
4	58	6	36	50	-	-	5	45
12	68	11	21	35	-	1	-	64
14	73	8	19	15	-	5	-	80

H.M. = Heavy Mineral.

Table 4. Radionuclide Sorption Ratio (Kd) of Soil Samples from Beatty and Barnwell Low-Level Radioactive Waste Sites in Normal and Controlled Atmospheres

Sample Number	Radionuclides						
	U	Sr	Ni	Co	Cs	Pu	Fe
Radionuclide Sorption Ratio (Kd) - Oxic Conditions							
Beatty	1	100	2,100	2,600	3,300	700	1,200
	2	82	3,200	620	4,500	5,000	6,000
	5	150	3,600	9,000	8,200	1,800	1,800
Barnwell	4	82	115	96	3,500	22,500	6,000
	12	142	123	136	3,100	3,700	6,000
	14	190	116	24	1,500	135	600
Kd - Controlled Atmosphere - Reducing Conditions							
Beatty	1	157	1,700	1,800	4,300	16,000	800
Barnwell	14	115	150	130	900	120,000	680
							140,000
							120,000

Preliminary evaluation of the West Valley, New York, site samples reveals a texture and composition relationship in which clay minerals (illite and chlorite) are contained predominantly in the clay-size fraction. Thus, for two of the three major surficial deposits, the clay minerals are predominantly in the clay-size fraction. The exception was the occurrence of kaolinite at the Barnwell site in both the silt and clay-size fractions.

SORPTION COEFFICIENT VERSUS PERCENT SORPTIVE MINERALS

Most of the radionuclides at the Barnwell and Beatty sites show a general increase of sorption coefficient (K_d) with increased sorptive minerals. A linear relationship was found limited to cesium and strontium for the Beatty site and for strontium at the Barnwell site (Figure 7). Daniels, et al. (1982), found a similar relationship for cesium and strontium in relation to clinoptilolite from the Yucca Mountain (Nevada) tuff samples. They attribute the proportional K_d relationship to the presence of clinoptilolite since the K_d of smectite (montmorillonite) did not follow this relationship. The range of K_d values found for clinoptilolite in the Yucca Mountain tuff samples are generally similar for percentages of sorptive minerals in the 10 percent range such as occurs for the Beatty site samples. Daniels, et al. (1982), using a nonweighted least squares fit to data points in which clinoptilolite was approximately 10 percent, found K_d values for strontium and cesium of 690 ± 170 and 430 ± 150 , respectively. The K_d values for strontium and cesium at the Beatty site are 80 to 150 and 3000 to 8000, respectively (Figure 7). However, the clinoptilolite approximates 3 percent and montmorillonite 7 percent of the Beatty samples. Considering that montmorillonite has a K_d of 4900 for cesium and 150 for strontium (Ames and Rai, 1979), it would appear that at the Beatty site the montmorillonite contributes to the sorption ratio.

At the Barnwell site, the plot of percent sorptive kaolinite versus K_d of strontium provides a straight-line relationship as depicted in Figure 7. This same relationship is not apparent for cesium at the Barnwell site as it is at the Beatty site. Some of the data, however, is still in doubt and additional data are being obtained.

OTHER CONTROL FACTORS

In the more permeable media at the Beatty and Barnwell sites, oxygenated conditions prevail in the trenches. However, because of the waste form and impervious nature of the soil of the West Valley site, reducing conditions occur in some of the trenches (Weiss and Colombo, 1979). The sorptive behavior of radionuclides for site-specific soils at the Beatty and Barnwell sites was tested by the Los Alamos Laboratory using a controlled atmosphere of nitrogen (less than 0.2 ppm oxygen and less than 20 ppm carbon dioxide) to determine if sorptive behavior of some of the radionuclides may be different from that occurring under normal atmospheric conditions. The K_d values under the controlled

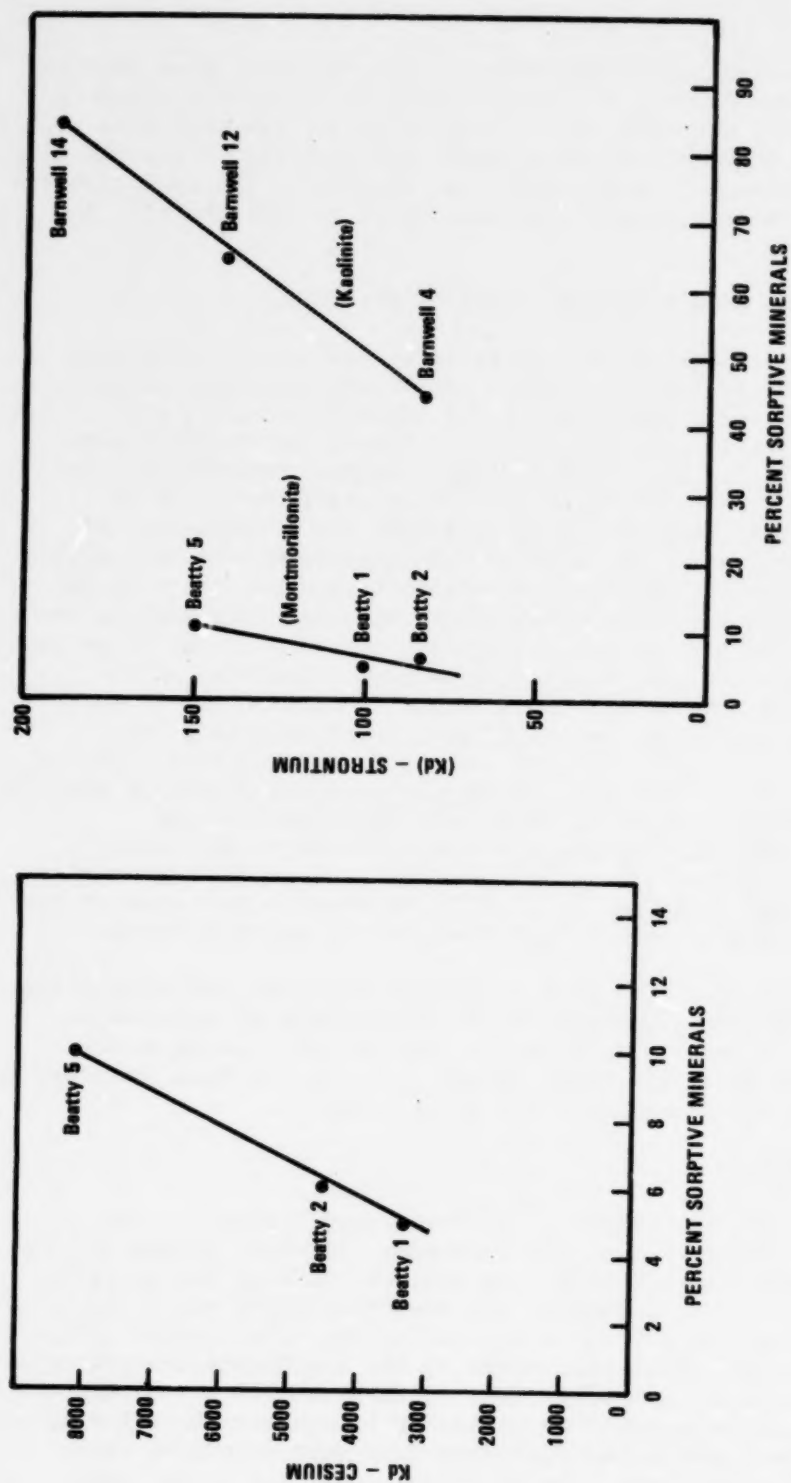


Figure 7. Percent sorptive minerals versus sorption coefficient (K_d) of cesium and strontium for samples from the Barnwell and Beatty low-level radioactive waste sites.

reducing atmospheric conditions are listed in Table 4. The sorption of plutonium under reducing conditions is higher by a factor of 3 for the Beatty sample and by a factor of 6 for the Barnwell sample (Wolfsberg, et al., 1983). Uranium was not tested in the controlled atmosphere. Similar Kd values were obtained for strontium, cesium, iron, nickel, cobalt, and americium in both ambient and controlled atmospheres.

The differences in the Kd values of nickel and cobalt at the LLW sites are believed to be mainly from chemical control factors. The Beatty site Kd values are an order-of-magnitude higher than those at the Barnwell or West Valley sites. Swanson (1982) showed that by changing the pH of an alkaline Hanford solid to an acidic condition (pH-6.0) with other factors similar, the Kd of nickel and cobalt would be reduced tenfold. Jenne (1968) presents evidence that the principal sorption mechanism of cobalt and nickel in the marine sediments is the coprecipitation of these radionuclides with hydrous oxides of manganese and iron under controlling conditions of pH and Eh (redox potential). Similar chemical factors probably control the high retention that might be expected for cobalt and nickel in the Alluvial Basin deposits relative to the lower retention in the Glaciated Central Region and Coastal Plain deposits. More work needs to be done to explore the effects of other factors before prediction parameters are feasible for nickel or cobalt.

The low sorption of uranium by the Beatty samples compared to that of the Barnwell samples may be related to the relatively high concentrations of bicarbonate at Beatty that give rise to poor sorbing anionic complexes. Similarly, the americium and plutonium colloid chemistries may be affected by the groundwater composition (Wolfsberg, et al., 1983).

SUMMARY AND CONCLUSIONS

Each of the low-level radioactive waste sites investigated for predictive parameters to assess radionuclide retention is situated in contrasting surficial deposits of the contiguous United States. Each of these surficial deposits has a unique assemblage of minerals, texture, and chemical environmental factors that are products of the climate and geologic history of the deposits. A comparison of the sorption coefficients (Kd's) for radionuclides at the three sites reveals repeated similarities within each deposit but large variations in the Kd values between the contrasting surficial deposits. The Alluvial Basin deposit has higher retention of radionuclides than either the Glaciated Central Region deposit or the Coastal Plain deposits.

The most obvious differences in the surficial deposits are (1) the type and amount of sorptive minerals and (2) the acidic or alkaline pH. The sorptive minerals are also the finest textured components of the surficial deposits. Quartz and other minerals common to the sand- and silt-size particles generally have low sorptive properties.

An assessment of sorptive minerals using established methods to focus on their quantitative proportions in representative and fractionated samples provides a critical parameter for comparing measured sorption coefficient (K_d) values of radionuclides. While retention of most radionuclides increases with increased amounts of sorptive minerals, a linear relationship between K_d and sorptive minerals is evident for only cesium and strontium. Linear relationships of percent sorptive minerals versus sorption coefficient (K_d) values are apparent in the preliminary evaluations of the Alluvial Basin deposits for both cesium and strontium and in the Coastal Plain deposits for strontium. The K_d determination of the Glaciated Central Region deposits is not yet completed; however, it becomes apparent from this preliminary investigation that cesium and strontium may be predictable by careful evaluation of the sorptive minerals in the potential low-level radioactive waste sites of each region.

The alkaline nature of the Alluvial Basin deposits appears to account for an order-of-magnitude higher retention of cobalt and nickel in these deposits than in the acidic surficial deposits. Other chemical factors, such as presence or absence of iron, manganese, and organics, and coprecipitation with a hydrous oxide, may also relate to the retention of cobalt and nickel.

The groundwater chemistry may be significant in controlling retention of uranium, americium, and plutonium in the surficial deposits. Higher uranium retention in the Barnwell deposits may be related to low bicarbonate concentration as contrasted to the low uranium retention in the other two deposits containing greater amounts of this ligand.

The sorption coefficients (K_d 's) of tritium, carbon-14, technetium, and iodine are predictable in that there is negligible retention of these radionuclides in any of the surficial deposits due to their occurrence as tritium in water or a covalently bonded compound such as C-14 or as anions.

RECOMMENDATIONS

Linear relationships that may exist as predictive parameters of the major surficial deposits can be determined by testing site-specific soils for the sorption coefficient (K_d) of radionuclides and the percent and variety of sorptive minerals by using a quantitative mineral evaluation method. Variations of amounts of sorptive minerals by fractionation techniques must maintain the chemical integrity of all samples. Verification for regional consistency should be conducted on a number of samples within similar surficial deposits to validate the methodology apparent in this investigation.

Chemical factors which are important as control mechanisms are not as easily identified as predictive parameters for radionuclide retention. Comparison of the radionuclide retention between major surficial deposits, however, are suggestive of potential control factors. More investigations identifying limits and ranges of chemical factors for several groundwaters with variation in radionuclide concentrations are required to provide information applicable to prediction of radionuclide retention with chemical parameters at this time.

It is recommended that efforts continue to provide data on radionuclide retention capabilities for the value it will afford assessment of future low-level radioactive waste sites. Preliminary predictive data realized in use of texture and quantitative mineralogical techniques may eventually also encompass chemical parameters. The benefits realized will be reflected in health benefits to the nation as well as advancing our understanding of physicochemical factors relating to protection of our groundwater from pollution.

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